Linear response formula for open systems

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An exact expression for the finite frequency response of open classical systems coupled to reservoirs is obtained. The result is valid for any conserved current. No assumption is made about the reservoirs apart from thermodynamic equilibrium. At non-zero frequencies, the expression involves correlation functions of boundary currents and cannot be put in the standard Green-Kubo form involving currents inside the system.

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I. INTRODUCTION

One of the important tools in the study of transport phenomena is the Green-Kubo formula[1, 2], which relates the equilibrium correlation function of any two conserved currents (i.e. currents associated with conserved charges) to the linear response of the first when a gradient in the potential conjugate to the second is applied. The response is expressed in terms of conductivity coefficients of a system, in the thermodynamic limit when boundary effects can be neglected. The proof[2] relies on Onsager's[3] relation between the time evolution of equilibrium and non-equilibrium fluctuations, or obtains the response to an external field[4] and relates it to the response to an internal gradient[5]. One can thence obtain the conductance of a system if it is sufficiently large and if the conductivity does not diverge.

If either of these conditions breaks down, one has to find the conductance of the system directly. This can be done by connecting it to infinite reservoirs[6, 7], but requires assumptions about the reservoirs and the appropriate ones can be subtle[8]. Alternatively, a Green-Kubo like formula has been obtained[9] for the conductance(s) of an open classical system without having to deal with the reservoirs. This is done by assuming that the system is in a non-equilibrium steady state and (implicitly) that the current driven by the external potential gradient is a scalar. The assumptions restrict the result to the zero-frequency conductance of a quasi one-dimensional (in a sense to be made precise later) system.

In this paper, we derive a formula for the generalized linear conductance of a finite classical system without any of these restrictions. The system can have an arbitrary shape and number of reservoirs, and the formula applies at any frequency. The only assumption made about the reservoirs is that they are in thermodynamic equilibrium. The coupling between the system and the reservoirs is assumed to be such that if the system starts in equilibrium at the same thermodynamic potentials as all the reservoirs, it remains in equilibrium, but we do not assume that the reservoirs can equilibrate the system. For a quasi one-dimensional system at zero frequency, the formula can be transformed into that of Ref. [9] which is of the standard Green-Kubo form, i.e. it involves the equilibrium fluctuations of currents inside the system. We show that this transformation is not possible at finite frequencies.

The derivation here builds on previous work that obtained the thermal conductance at zero[10] and non-zero[11] frequency of a finite classical system in contact with heat baths. Although a large variety of heat baths were considered, the proof had to be painstakingly constructed separately for each bath. Thus it was not clear whether it might fail for some types of heat baths and whether it applied to other conserved currents. The present paper resolves these questions.

The rest of this paper is organized as follows. In Section II , we briefly review the result of Ref. [10, 11] for the thermal conductance of a Fermi-Pasta-Ulam (FPU) chain with Langevin heat baths at the ends. The notation is more general and part of the proof is slightly different to allow it to be extended to other conserved currents. In Section III, we extend the proof to the case of all conserved currents. In Section IV we transform the formula at zero frequencies to the 'standard' form, and discuss why this is not possible at non-zero frequencies.

II. FPU CHAINS WITH LANGEVIN BATHS

We first review the derivation of Refs. [10, 11] for the heat conductance of a N-particle FPU chain with Langevin baths at the ends. The equations of motion are

$$
\begin{aligned}
m_l \dot{v}_l = & -\frac{\partial}{\partial x_l}[U(x_{l-1} - x_l) + U(x_l - x_{l+1}) + V(x_l)] \\
& + \delta_{l,1}[\eta_L(t) - \gamma_L v_1] + \delta_{l,N}[\eta_R(t) - \gamma_R v_N] \quad (1)
\end{aligned}
$$

for $l = 1, 2, \ldots N$. Here m_l, x_l, v_l are the mass, position and velocity of the l'th particle. U and V are the interparticle and onsite potentials with $x_0 = x_{N+1} = 0$. $\gamma_{L,R}$ and $\eta_{L,R}(t)$ are the damping and Gaussian noise from reservoirs at temperatures $T_{L,R}$, satisfying

$$
\langle \eta_{L,R}(t)\eta_{L,R}(t') \rangle = 2\gamma_{L,R}k_B T_{L,R}\delta(t - t'). \quad (2)
$$

In the first stage of the proof, the Fokker Planck equation for the full phase space distribution function $P(\mathbf{x}; \mathbf{v}; t)$ is constructed, where $\mathbf{x} = \{x_1 \ldots x_N\}$ and $\mathbf{v} = \{v_1 \ldots v_N\}$. If $T_L = T_R$, the steady state solution to

the equation is the equilibrium Boltzmann distribution $P^0(\mathbf{x}, \mathbf{v})$. For $T_{L,R} = T \pm \frac{1}{2}\Delta T$, we have

$$\frac{\partial P}{\partial t} == \hat{L}P + \hat{L}^{\Delta T}P \qquad (3)$$

where $\hat{L}$ is the equilibrium ($\Delta T = 0$) Fokker Planck operator

$$\hat{L} = \frac{\gamma_L}{m_1}\left[\frac{k_B T}{m_1}\frac{\partial^2}{\partial v_1^2} + \frac{\partial}{\partial v_1}v_1\right] + (1, L) \to (N, R)$$
$$+ \sum_i \left[-v_i\frac{\partial}{\partial x_i} - F_i\frac{\partial}{\partial v_i}\right] \qquad (4)$$

(F_i is the force on the i'th particle) and

$$\hat{L}^{\Delta T} = \frac{k_B \Delta T}{2}\left[\frac{\gamma_L}{m_1^2}\frac{\partial^2}{\partial v_1^2} - \frac{\gamma_R}{m_N^2}\frac{\partial^2}{\partial v_N^2}\right]. \qquad (5)$$

With $P(\mathbf{x}, \mathbf{v}, t) = P^0 + p(\mathbf{x}, \mathbf{v}, t)$, to linear order in ΔT

$$p(\mathbf{x}; \mathbf{v}; t) = \int_{-\infty}^t e^{(t-t')\hat{L}} J_{fp}^e(\mathbf{v})P^0(\mathbf{x}, \mathbf{v})\Delta\beta(t')dt' \quad (6)$$

where $\Delta\beta = \Delta(1/k_B T) = -\Delta T/(k_B T^2)$ and $J_{fp}^e(\mathbf{v})$ is defined by

$$\left.\frac{\partial P}{\partial t}\right|_{P=P^0} = \hat{L}^{\Delta T}P^0 = (\Delta\beta)J_{fp}^e P^0 \qquad (7)$$

(the superscript in J_{fp}^e referring to the energy). For any observable A, we define $\langle\delta A\rangle = \langle A\rangle - \langle A\rangle_0$ in terms of its expectation values with $\Delta\beta \neq 0$ and $\Delta\beta = 0$. Then

$$\langle\delta A(t_1)\rangle_{\Delta T} = \int_{-\infty}^{t_1} \langle A(t_1)J_{fp}^e(t_2)\rangle_{eq}\Delta\beta(t_2)dt_2 \qquad (8)$$

where the correlation function on the right hand side is evaluated in equilibrium with $\Delta T = 0$.

More generally, if the reservoirs at the ends have different values for some thermodynamic potential Φ^ρ whose conjugate conserved density is ρ, then

$$\langle\delta A(t_1)\rangle_{\Delta\Phi} = \int_{-\infty}^{t_1} \langle A(t_1)J_{fp}^\rho(t_2)\rangle_{eq}\Delta\Phi^\rho(t_2)dt_2 \qquad (9)$$

with J_{fp}^ρ defined by the generalization of Eqs.(3) and (7). For example, for the particle current, $\Phi^n = -\beta\mu$, where μ is the chemical potential.

The second part of the proof is specific to Langevin baths at different temperatures, and is slightly different from Refs. [10, 11]. From the Fokker Planck equation, one can verify that

$$J_{fp}^e = \frac{\gamma_R}{2m_N}[m_N v_N^2 - k_B T] - \frac{\gamma_L}{2m_1}[m_1 v_1^2 - k_B T]. \quad (10)$$

We define the boundary energy current variable J_b^e as the mean of the instantaneous energy currents flowing into

the system from the left reservoir and flowing out of the system to the right reservoir. Thus

$$J_b^e(t) = \frac{1}{2}(j_{1,L}^e - j_{N,R}^e) \qquad (11)$$

where

$$j_{1,L}^e(t) = -\gamma_L v_1^2(t) + \eta_L(t)v_1(t),$$
$$j_{N,R}^e(t) = -\gamma_R v_N^2(t) + \eta_R(t)v_N(t). \qquad (12)$$

Then we will prove the relation

$$\langle A(t_1)J_{fp}^e(t_2)\rangle_{eq} = -\langle A(t_1)J_b^e(t_2)\rangle_{eq} \qquad \text{if } t_1 > t_2. \qquad (13)$$

To prove Eq.(13), we use Eq.(12) and average over the noise. In performing this average, we discretize the equations of motion Eq.(1) in the usual manner, with the right hand side evaluated at time t and the left hand side equal to $m_l[v_l(t+\epsilon) - v_l(t)]/\epsilon$. The boundary heat currents are

$$j_{1,L}^e(t) \equiv \frac{1}{2}[v_1(t) + v_1(t+\epsilon)][\eta_L(t) - \gamma_L v_1(t)]$$
$$j_{N,R}^e(t) \equiv \frac{1}{2}[v_N(t) + v_N(t+\epsilon)][\eta_R(t) - \gamma_R v_N(t)] (14)$$

Usually, the noise average is performed at fixed $v_1(t)$, yielding

$$\langle j_{1,L}^e(t)\rangle_\eta = \gamma_L(k_B T/m_1 - v_1^2(t)). \qquad (15)$$

Naively, this would seem to yield Eq.(13) without the minus sign on the right hand side.

However, we want to use the Fokker-Planck evolution operator over the interval $t_1 > t > t_2$ to evaluate the left hand side of Eq.(13). Therefore, we use $J_b^e(t_2 - \epsilon)$ in Eq.(13) and perform the noise average for fixed $v_{1,N}(t_2)$, not $v_{1,N}(t_2 - \epsilon)$. We change variables in Eq.(14) from $\eta_{L,R}(t)$ to $\tilde{\eta}_{L,R}(t) = \eta_{L,R}(t) - 2\gamma_{L,R}v_{1,N}$, since $\eta_{L,R}(t)$ are uncorrelated with $v_{1,N}(t+\epsilon)$. It is easy to verify that

$$\langle j_{1,L}^e(t-\epsilon)\rangle_\eta = \gamma_L(v_1^2(t) - k_B T/m_1),$$
$$\langle j_{N,R}^e(t-\epsilon)\rangle_\eta = \gamma_R(v_N^2(t) - k_B T/m_N). \qquad (16)$$

The same result can also be obtained by using the fact that evolving backwards in time from t_2 reverses the currents, and from the time reversal invariance of the dynamics the noise-averaged time reversed current is given by Eq.(15). Comparing with Eq.(7), the noise averaged $J_b^e(t_2 - \epsilon)$ is equal to $-J_{fp}^e(t_2)$, thus proving Eq.(13). Combining Eqs.(9) and (13), we have

$$\langle\delta A(t_1)\rangle_{\Delta T} = -\int_{-\infty}^{t_1} \langle A(t_1)J^e b(t_2)\rangle_{eq}\Delta\beta(t_2)dt_2 \quad (17)$$

which gives the response to a general time dependent (i.e. non-zero frequency) variation in the temperatures of the reservoirs. Equivalently, Fourier transforming,

$$\langle\delta A(\omega)\rangle_{\Delta T} = -\Delta\beta(\omega)\left[\int_0^\infty \exp[i\omega t]\langle A(t)J_b^e(0)\rangle_{eq}dt\right]. \qquad (18)$$

This equation is now generalized to all conserved currents in Section III.

III. GENERALIZED CURRENTS

We first consider the case of particle currents. Although Section II was for a lattice system with a fixed number of particles, it can be easily extended to a system with a continuous coordinate x in which the particles move around, allowing particles to be enter and leave the system from the reservoirs.

Because the number of particles in the system is no longer conserved, we work in the grand canonical ensemble. Let $P_N(\mathbf{x}, \mathbf{v}, t)$ be the probability density for the system to be in an N-particle configuration with coordinates $(\mathbf{x}, \mathbf{v})$ at time t. Thus $\int P_N(\mathbf{x}, \mathbf{v}, t) d\mathbf{x} d\mathbf{v}$ is the probability for the system to be in an N-particle configuration. In equilibrium, $\int P_N^0(\mathbf{x}, \mathbf{v}) d\mathbf{x} d\mathbf{v} = \exp[\beta\mu N] Z_N / \Omega$, where Z_N is the canonical partition function for the N-particle system and Ω is the grand partition function. The Fokker Planck probability density is now an infinite column vector, and the time evolution operators $\hat{L}$ and $\hat{L}^{\Delta\mu}$ are matrices. We will denote the column vector as $P(\mathbf{x}, \mathbf{v}, t)$. Analogous to Eq.(7), we define J_{fp}^n through

$$\left.\frac{\partial P}{\partial t}\right|_{P=P^0} = \hat{L}^{\Delta\mu} P^0 = (-\Delta\beta\mu) J_{fp}^n P^0. \qquad (19)$$

The operator $\hat{L}^{\Delta\mu}$ is due to the difference in the chemical potentials of the reservoirs and the system, and changes the number of particles N.

Let $\lambda_{N',N}^L(\beta\mu_L)$ be the transition rate from $P_N(x_1 \ldots v_N)$ to $P_{N'}(x'_1 \ldots v'_{N'})$ due to the left reservoir. (The dependence of λ on other variables such as $\mathbf{x}, \mathbf{v}$ and t is implicit; if some of the reservoir variables are so slow that they cannot be integrated out to yield transition rates, one can augment the arguments of P_N and the λ's to include these variables.) The contribution from the left reservoir to the first order equation for $\Delta P_N = P_N - P_N^0$ is

$$\partial_t^L \Delta P_N(\mathbf{x}_1 \ldots \mathbf{v}_N) = \Delta(\beta\mu)_L \sum_{N'} \int \left[(\partial_{\beta\mu_L} \lambda_{N,N'}^L) P_{N'}^0 \right.$$
$$\left. - (\partial_{\beta\mu_L} \lambda_{N',N}^L) P_N^0 \right] d\mathbf{x}'_1 \ldots d\mathbf{v}'_{N'} (20)$$

where the derivatives on the right hand side are taken at $(\beta\mu)_L = \beta\mu$. Here $\partial_t^L \Delta P_N$ is the part of the time evolution of ΔP_N due to the left reservoir; $\partial_t P_N = (\partial_t^L + \partial_t^R) P_N$.

Since the left reservoir cannot disturb the grand canonical ensemble distribution when it is at the same $\beta\mu$ as the system, we have

$$\sum_{N'} \int \lambda_{N,N'}^L P_{N'}^0 d\mathbf{x}'_1 \ldots d\mathbf{v}'_{N'} = \sum_{N'} \int \lambda_{N',N}^L P_N^0 d\mathbf{x}'_1 \ldots d\mathbf{v}'_{N'}$$
$$(21)$$

whenever $(\beta\mu)_L = \beta\mu$. The derivatives on the right hand

side of Eq.(20) can then be transferred:

$$\partial_t^L \Delta P_N(\mathbf{x}_1 \ldots \mathbf{v}_N) = -\Delta(\beta\mu)_L \sum_{N'} \int \left[\lambda_{N,N'}^L (\partial_{\beta\mu} P_{N'}^0) \right.$$
$$\left. - \lambda_{N',N}^L (\partial_{\beta\mu} P_N^0) \right] d\mathbf{x}'_1 \ldots d\mathbf{v}'_{N'} (22)$$

Since $P_N^0 \sim \exp[N\beta\mu]$, applying Eq.(21) again

$$\partial_t^L \Delta P_N = \Delta(\beta\mu)_L \sum_{N'} (N - N') \int \lambda_{N,N'}^L P_{N'}^0. \qquad (23)$$

Now $\lambda_{N,N'}^L P_{N'}^0$ integrated over $\mathbf{x}'_1 \ldots \mathbf{v}'_{N'}$ is the rate at which systems transition from an N' particle state to an N-particle state at the phase space point $\mathbf{x}_1 \ldots \mathbf{v}_N$. Therefore the probability density for the system to be in an N-particle state at time t and have received $N - N'$ particles from the left reservoir between time $t - \delta t$ and t is $\lambda_{N,N'}^L P_{N'}^0 \delta t$. For a system in an N-particle state at $\mathbf{x}_1 \ldots \mathbf{v}_N$, the particle current from the left reservoir immediately *before* the time t is then $j_{1,L}^N P_0^N(\mathbf{x}, \mathbf{v}) = \sum_{N'} (N - N') \lambda_{N,N'}^L P_{N'}^0$. The stipulation that the current has to be evaluated just before t is important as seen from the discussion around Eqs.(15) and (16).

Comparing with Eq.(23), including the effect of the reservoir to the right with $\Delta(\beta\mu)_L = -\Delta(\beta\mu)_R = \frac{1}{2}\Delta(\beta\mu)$ and using Eq.(19), we obtain

$$J_{fp}^n P^0 = -J_b^n P^0 \qquad (24)$$

where J_b^n is defined in terms of the particle currents from the reservoirs in the same manner as Eq.(11).

Although energy currents were handled differently in Section II, there is no reason why one could not have proceeded as we have done here: define $P_E(\mathbf{x}, \mathbf{v}, t)$ as the probability density for the system to have an energy E and be at the phase-space point $(\mathbf{x}, \mathbf{v})$. (The sums over N' would have been replaced by integrals over E'.) Since the number of variables in $(\mathbf{x}, \mathbf{v})$ is independent of the energy E and different energies correspond to non-overlapping regions in phase space, this would have been an unnecessary complication. But from this we see that although the specific example of particle currents has been used in this Section, *all* conserved currents can be treated in the same manner, and

$$\langle A(t_1) J_{fp}^\rho(t_2) \rangle_{eq} = -\langle A(t_1) J_b^\rho(t_2) \rangle_{eq} \qquad \text{if } t_1 > t_2 \quad (25)$$

for any conserved current J^ρ. Nor is one limited to two reservoirs: since the proof we have constructed for Eq.(25) deals with the reservoirs separately, one can consider an arbitrary number of reservoirs at potentials Φ_α^ρ. Combining with Eq.(9),

$$\langle \delta A(t) \rangle_{\Delta\Phi} = \sum_\alpha \int_{-\infty}^t \langle A(t) j_{b,\alpha}^\rho(t') \rangle_{eq} \Delta\Phi_\alpha^\rho(t') \, dt' \qquad (26)$$

If A is the current $j_{b,\kappa}^{\sigma}$ at the boundary κ, one obtains the conductance [12]:

$$\langle j_{b,\kappa}^{\sigma}(t)\rangle_{\Delta\Phi} = \sum_{\alpha,\rho}\int_{-\infty}^{t}\langle j_{b,\kappa}^{\sigma}(t)j_{b,\alpha}^{\rho}(t')\rangle_{eq}\Delta\Phi_{\alpha}^{\rho}(t')\,dt'.$$
(27)

As with Eq.(17) this can be Fourier transformed to yield

$$\langle j_{b,\kappa}^{\sigma}(\omega)\rangle_{\Delta\Phi} = \sum_{\alpha,\rho}\Delta\Phi_{\alpha}(\omega)\int_{0}^{\infty}\exp[i\omega t]\langle j_{b,\kappa}^{\sigma}(t)j_{b,\alpha}^{\rho}(0)\rangle_{eq}dt.$$
(28)

The equilibrium correlation function in Eq.(26) must be evaluated with the *same* boundary conditions as the left hand side, i.e. with reservoirs connected to the open system (but, unlike the left hand side, with the reservoirs at the same thermodynamic potentials). Different boundary conditions may be inequivalent even in the large system limit [11, 13].

IV. ZERO FREQUENCY CONDUCTANCE

At zero frequencies, for a quasi one dimensional system with two reservoirs, Eq.(9) can be transformed into a more familiar form. The proof is a generalization of the one in Ref. [10]. If the reservoirs are at potentials $\Phi^{\rho}\pm(\Delta\Phi^{\rho})/2$, from Eqs.(9) and (25)

$$\langle\delta A(t)\rangle_{\Delta\Phi} = \sum_{\rho}(\Delta\Phi^{\rho})\int_{-\infty}^{t}\langle A(t)J_{b}^{\rho}(t')\rangle_{eq}dt'. \quad (29)$$

For concreteness, we first consider a one dimensional lattice. Let ρ_i be the amount of the conserved quantity ρ at the i'th particle, $j_{i+1,i}^{\rho}$ be its current between the i'th and $i+1$'th particles, and $J^{\rho}(t) = \sum_{i=0}^{N}j_{i+1,i}^{\rho}(t)$. If $A = J^{\sigma}$ is the current flowing through the entire chain that is associated with a conserved quantity σ, Eq.(29) becomes

$$\langle\delta J^{\sigma}(t)\rangle_{\Delta\Phi} = \sum_{\rho}(\Delta\Phi^{\rho})\int_{-\infty}^{t}\langle J^{\sigma}(t)J_{b}^{\rho}(t')\rangle_{eq}dt'. \quad (30)$$

Define $D^{\rho}(t) = \sum_{k=1}^{N}(N+1-2k)\rho_k$. From the continuity equation,

$$dD^{\rho}/dt = -2J^{\rho}(t) + 2(N-1)J_{b}^{\rho}(t). \quad (31)$$

Multiply both sides of this equation by $J^{\sigma}(t_1)$, take an equilibrium average, and integrate over $-\infty < t < t_1$. Then $\langle J^{\sigma}(t)D^{\rho}(-\infty)\rangle_{eq} = \langle J^{\sigma}\rangle_{eq}\langle D^{\rho}\rangle_{eq} = 0$, and if ρ and σ have the same symmetry under time reversal or the system is symmetric under reflection about its middle,

$$\langle J^{\sigma}(t)D^{\rho}(t)\rangle_{eq} = 0. \quad (32)$$

If either condition is satisfied for each ρ in Eq.(30),

$$\langle\delta J^{\sigma}(t)\rangle_{\Delta\Phi} = \frac{1}{N-1}\sum_{\rho}(\Delta\Phi^{\rho})\int_{-\infty}^{t}\langle J^{\sigma}(t)J^{\rho}(t')\rangle_{eq}dt'.$$
(33)

For a continuum system of length L, D^{ρ} is defined as $\int\rho(x)(L-2x)dx$, and Eqs.(31) and (33) are obtained with $N-1$ replaced by L. The factor of $N-1$ in Eqs.(31) and (33) is replaced with L. In higher dimensions, we define $D^{\rho} = -2\int x\rho(\mathbf{r})d\mathbf{r}$ and use

$$dD^{\rho}/dt = 2\int x\nabla\cdot j^{\rho}(\mathbf{r})d\mathbf{r} = -2\int j_x^{\rho}(\mathbf{r}) + 2\sum_{\alpha=1,2}j_{b,\alpha}^{\rho}x_{\alpha}$$
(34)

where j_x is the x component of the current and the sum is over the reservoirs. Then if Eq.(32) is satisfied, Eq.(33) is obtained with $J^{\rho}\to J_x^{\rho}$ on the right hand side and $N-1$ replaced by the x-separation of the two reservoirs[14].

Eq.(32) is always satisfied for quasi one dimensional homogeneous systems. By quasi one dimensional, we mean that the system is a tube whose cross-section does not vary along its length, with reservoirs at its ends. Translational invariance within the tube ensures that currents in response to potential differences between the reservoirs are parallel to the orientation of the tube, and so are effectively scalar. This is implicitly assumed in Ref. [9], which derives Eq.(33) for a non-equilibrium steady state. Eq.(32) is also satisfied if $\rho = \sigma$.

Since both particle and energy currents change sign under time reversal, the continuum version of Eq.(33) is valid for thermoelectric transport coefficients:

$$\begin{aligned}\overline{j}^{n} &= G^{nn}\Delta(\beta\mu) - G^{ne}\Delta\beta\\ \overline{j}^{e} &= G^{en}\Delta(\beta\mu) - G^{ee}\Delta\beta\end{aligned} \quad (35)$$

where $\overline{j}^{n}, \overline{j}^{e}$ are non-equilibrium steady state currents flowing in response to differences in the chemical potentials and temperatures of the reservoirs, and $G^{\sigma\rho} = \int_0^{\infty}\langle j^{\sigma}(t)j^{\rho}(0)\rangle dt$.

Despite the similarity between Eq.(33) and the Kubo formula for the conductivity [2, 4], there are important differences. The caveat at the end of Section III is still valid: the correlation function has to be calculated with the correct boundary conditions. Second, Eq.(33) has *only* been obtained in the zero frequency limit.

One might wonder whether the $\omega\neq 0$ version of Eq.(33), with $\Delta\Phi^{\rho}(t')$ taken inside the integral, might be true even if not proved here. A simple example proves otherwise: if the contacts to the reservoirs are so weak that the system is effectively isolated, a non-zero $\Delta\Phi$ produces no response and the boundary current $j_{b,\alpha}^{\rho}$ in Eq.(26) is zero. However, $\langle J^{\sigma}(t)J^{\rho}(t')_{eq}\rangle\neq 0$ because currents flow inside an isolated system due to spontaneous fluctuations, and $\int_{-\infty}^{t}\langle J^{\sigma}(t)J^{\rho}(t')_{eq}\Delta\Phi^{\rho}(t')dt'\neq 0$. By contrast, if the system is driven by an applied field instead of a difference in potentials between the reservoirs, the $\omega\neq 0$ version of Eq.(33) is always valid.

To summarize, we have obtained a formula for the generalized finite-frequency response of a finite classical system connected to an arbitrary number of reservoirs when the thermodynamic potentials of the reservoirs are varied. The equilibrium correlation function in the formula involves currents flowing in from the reservoirs. At zero

frequency, it reduces to a familiar Green-Kubo form, i.e. it involves equilibrium fluctuations of the currents inside the system but this is not the case at finite frequencies.

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